

## REMARKS

The claims are 2 to 4.

Favorable reconsideration is respectfully requested.

New claim 2 is independent with new claims 3 and 4 dependent thereon. The significance of these claims will be discussed below.

The specification and abstract have been reviewed and revised, and a substitute specification and abstract has been prepared. No new matter has been added.

Also enclosed is a "marked-up" copy of the original specification and abstract to show the changes that have been incorporated into the substitute specification and abstract. The enclosed copy is entitled "Version with Markings to Show Changes Made".

The above amendment is responsive to points set forth in the Official Action.

In Official Action page 2, with regard to the rejection under 35 U.S.C. 112 on the ground that the claim is incomplete, there is now recited a heating step during which the volume of the gallium is expanded, as well as a step in which the gallium is oxidized in the presence of air to produce gallium oxide which is firmly bonded to the nanotube.

Thus, the rejection under 35 U.S.C. 112 is inapplicable to the present claims.

Claim 1 has been rejected under 35 U.S.C. 102(a) as anticipated by Gao.

This rejection is respectfully traversed.

Gao discloses a carbon nanothermometer/method for measuring the temperature using a minute size temperature sensing element, as a temperature sensing element comprising a carbon nanotube with a continuous gallium column contained with one end opened and the other end closed.

The method of Gao comprises the steps of measuring the gallium length by a transmission electron microscopy at different temperature environments.

The environment is a vacuum to avoid mechanical damage to the transmission electron microscope. Thus, the gallium is not oxidized.

For this reason, the gallium cannot be bonded to the nanotube and the heated gallium contracts as it cools down.

In contrast, in the present claims gallium is expanded as in Gao but is oxidized and bonded to the carbon nanotube. The leading edge of the gallium oxide in the carbon nanotube is a function of the extent to which the gallium in the nanotube has expanded which in turn is a function of its temperature.

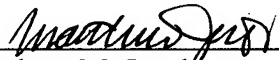
For the foregoing reasons, it is apparent that the rejection on Gao is untenable and should be withdrawn.

No further issues remaining, allowance of this application is respectfully requested.

If the Examiner has any comments or proposals for expediting prosecution, please contact undersigned at the telephone number below.

Respectfully submitted,

Yoshio BANDO et al.

By:   
Matthew M. Jacob  
Registration No. 25,154  
Attorney for Applicants

MJ/aas  
Washington, D.C. 20006-1021  
Telephone (202) 721-8200  
Facsimile (202) 721-8250  
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## SPECIFICATION

Version with Markings to  
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### METHOD FOR MEASURING TEMPERATURE USING MINUTE SIZE TEMPERATURE SENSING ELEMENT

#### Technical Field

The present invention relates to a temperature measuring method using a minute size temperature sensing element. More specifically, it relates to a novel temperature measuring method capable of measuring the temperature accurately in over a wide range, using a temperature sensing element comprising a carbon nanotube containing ~~a~~ columnar gallium.

#### Background Art

After the discovery of the carbon nanotube in 1991, a number of studies have ~~ever~~ been carried out by many researchers. ~~Then, various~~ Various technical ~~improvement and utilization~~ methods improvements and methods of utilization have been ~~found out~~ discovered concerning the carbon ~~nanotube~~ nanotubes. For example, nowadays, it is utilized widely for ~~the~~ field effect devices, ~~the~~ probe top ends for ~~the~~ scanning probe microscopes, ~~the~~ superconductive materials, ~~the~~ highly sensitive microbalances, ~~the~~ structural materials, ~~the~~ minute forceps for the nanoscale operation, ~~the~~ parts for ~~the~~ gas detectors, ~~the~~ hydrogen energy storage devices, or the like. Moreover, ~~the~~ studies have been executed actively for containing the various fillers in the carbon nanotube (Document 1 and Document 2).

For example, as the substances to be contained in the carbon nanotube, a metal such as ~~a~~ lead, ~~a~~ tin, ~~a~~ copper, ~~an~~ indium, ~~and a~~ mercury, an alkali metal such as ~~a~~ lithium, ~~a~~ sodium, ~~a~~ potassium, ~~a~~ rubidium, and ~~a~~ cesium, a superconductor such as ~~a~~ lead, ~~a~~ tin, and ~~a~~ gallium, a semiconductor such as ~~a~~ silicon, ~~a~~ germanium, ~~a~~ gallium arsenide, ~~a~~ zinc selenide, and ~~a~~ zinc sulfide, a magnetic material such as ~~a~~ samarium, ~~a~~ gadolinium, ~~a~~ lanthanum, ~~an~~ iron, ~~a~~ cobalt and ~~a~~ nickel, and a mixture thereof have been discussed.

Moreover, an organic molecule semiconductor such as a naphthalene, ~~an~~ anthracene, ~~a~~

phenanthrene, ~~a~~-pyrene, and ~~a~~-perylene, an organic dye molecule such as a cyanine dye and a  $\beta$ -carotene, and furthermore, a gas molecule such as ~~a~~-hydrogen fluoride, ~~a~~-methane, and ~~an~~-ethane, or the like have been discussed.

On the other hand, recently, a number of researchers have been involved in the study ~~field~~ of the micrometer size area so that the nano thermometer capable of measuring the temperature in the micrometer size environment has been ~~more and more~~ increasingly called for. However, the nano thermometers known so far have a relatively narrow measurable temperature range so that several kinds of the thermometers need to be prepared for each temperature range to be measured in the case of measuring the temperature ~~in~~ over a wide range. Due to ~~the~~ troublesomeness and ~~the~~ high cost, development of ~~the~~ a nano thermometer capable of measuring ~~the~~ temperature ~~in~~ over a wide range by itself has strongly been desired.

Under such circumstances, a nano thermometer utilizing a gallium, capable of accurately measuring the temperature ~~in~~ over a relatively wide temperature range has been proposed. As the principle of the temperature measurement, the linear expansion or contraction of the gallium according to the temperature change ~~in~~ over a wide range is utilized. By measuring the change of the length of the columnar gallium by a high resolution transmission electron microscopy, the temperature is measured.

~~Then, the~~ The production method for a temperature sensing element comprising a carbon nano tube having a 1 to 10  $\mu\text{m}$  length for containing the columnar gallium and a 40 to 150 nm diameter is known already (Document 3). Moreover, ~~by the present inventors~~ by the present inventors ~~[[,]]~~ the production method for a temperature sensing element according to the heat treatment at 1,200 to 1,400°C of gallium oxide powders and carbon powders under ~~the~~ inert gas flow has been developed by the present inventors and it is already ~~filed for the~~ the subject of a patent application (Application 1).

However, as to the temperature measuring method, utilizing a nano thermometer utilizing a gallium column discussed ~~so far~~ above, the length of the columnar gallium as the temperature sensing element cannot be read ~~out~~ unless the subject to be measured is introduced into the observation area of the high resolution transmission type electron microscope. On the other hand, once the temperature sensing element ~~is taken out from the~~ removed from the inside of the subject to the outside for the temperature

measurement, since the length of the columnar gallium returns to the length at ~~the~~ room temperature, ~~the~~ an accurate temperature of the subject at the high temperature cannot be known.

Document 1: P. Ajayan et al., Nature, vol. 361, page 333, 1993

Document 2: Official gazette of the Japanese Patent Application Laid Open No. 6-227806

Document 3: Gao, Y.H. & Bando Y., Nature, 415,599 (2002)

Application 1: Patent Application No. 2002-67661

Accordingly, an object of the present invention is to solve the above-mentioned problems by providing a novel method capable of accurately measuring the temperature of a subject to be measured at a high temperature even in the case where a columnar gallium temperature sensing element as a nano thermometer utilizing a gallium is ~~taken out~~removed from the inside of the subject to have the temperature measured ~~for the measurement at the~~ at room temperature.

#### Disclosure of Invention

In order to solve the above-mentioned problems, the present invention provides a method for measuring the temperature using a minute size temperature sensing element, as a temperature sensing element comprising a carbon nano tube with a continuous columnar gallium contained with one end opened and the other end closed, ~~characterized in~~ comprising the steps of measuring the gallium length by a transmission electron microscopy in different temperature environments, inserting the temperature sensing element into a subject to be measured in the air, and ~~taking out~~removing the temperature sensing element from the subject to be measured for measuring the gallium length by ~~the~~ transmission electron microscopy.

#### Brief Description of Drawings

FIG 1 is a high resolution transmission electron microscope image of a gallium-containing carbon nano tube temperature sensing element before heating, observed at 20°C.

FIG 2(A) is a high resolution transmission electron microscope image of the gallium-containing carbon nano tube temperature sensing element after heating to 358°C in ~~the~~ air, observed at 20°C, and FIG.

2(B) is a graph of an energy dispersive X-ray spectroscopy thereof.

FIG. 3 is a high resolution transmission electron microscope image of the gallium-containing carbon nano tube temperature sensing element at 440°C in ~~the~~ air.

FIG. 4 is a high resolution transmission electron microscope image of the gallium-containing carbon nano tube temperature sensing element after heating to 440°C in ~~the~~ air, observed at 20°C.

The numeral in the figures denotes the following.

1. gallium oxide thin layer

#### Best Mode for Carrying Out the Invention

The present invention has the above-mentioned characteristics. The embodiments thereof will be explained hereinafter.

According to the temperature measuring method of the present invention, first, a temperature sensing element comprising a carbon nano tube with one end opened and the other end closed, containing a continuous ~~columnar~~ column of gallium is introduced into the observation area of a high resolution transmission electron microscope maintained at different temperatures for measuring the length of the gallium at each temperature. Then, the temperature sensing element is inserted into a subject to have the temperature measured so as to be placed in a heated environment in the air atmosphere. According to the heating operation, the volume of the gallium is expanded as well as the top end part thereof is oxidized so as to produce a gallium oxide. Since the gallium oxide is bonded with the carbon nano tube firmly so that the gallium oxide at the top end part has its position fixed even when the temperature sensing element is taken out from the subject to be measured, the temperature sensing element is taken out from the subject to have the temperature measured after cooling for measuring the length of the gallium of the temperature sensing element again using the high resolution transmission electron microscope. Thereby, the temperature of the subject ~~to have the temperature measured~~ can be measured.

Accordingly, the nano thermometer of the present invention utilizes the expansion characteristics of the gallium present inside the carbon nano tube according to the temperature change so that in principle it is not different from a thermometer generally used ~~in general~~ for measuring the

expansion or contraction change of a mercury.

However, it differs therefrom in that the carbon nano tube has about a 1 to 10  $\mu\text{m}$  length and a 40 to 150 nm diameter for the measurement of the temperature in a micrometer size environment so that it is extremely minute compared with the generally used thermometer.~~used in general~~ Therefore, in order to measure the length of the gallium in the minute carbon nano tube, use of an optical instrument such as a high resolution transmission electron microscope is necessary.

According to the present invention, as the reason for selecting ~~the~~ gallium as the substance to be contained in the carbon nano tube, ~~the~~ gallium ~~as~~ has the widest liquid phase range (29.78 to 2,403 °C) among the metals so that it has the excellent characteristics of a low vapor pressure at a high temperature. Furthermore, since the ~~gallium has the volume of~~ gallium increases ~~increased~~ linearly according to the temperature rise in a 50 to 500°C temperature range, and furthermore, ~~it has the~~ since its volume is reduced linearly in the case of lowering the temperature, it is suitable as a thermometer required to measure the temperature ~~in over~~ over a wide temperature range.

For example, compared with the liquid phase range (-38.87 to 356.58°C) of ~~a~~ mercury used for ~~an~~ ordinary temperature measurement, it has a wide temperature measurement range ~~at a over~~ high temperature range, and thus the effect of using ~~the~~ gallium for the nano thermometer is apparent.

As to the carbon nano tube containing a gallium, it can be produced according to various kinds of methods including a known method explained as the conventional technique, a method proposed by the present inventors, ~~[[,]] et al.~~

~~Then, with~~ With reference to the examples, embodiments will be explained in further ~~details~~ detail.

## Examples

### <Example 1>

A temperature sensing element was produced according to the method disclosed in the above-mentioned document 3. The structure thereof was confirmed by a high resolution transmission electron microscopy with an X-ray energy diffusion spectrometer mounted. The temperature sensing

element was applied onto a grid for the observation by the high resolution transmission electron microscopy. Then, the temperature sensing element was observed by the high resolution transmission electron microscopy was maintained at 20°C and 58°C for measuring the height of the gallium.

FIG 1 is a high resolution transmission electron microscope image showing the height of the gallium at the time of observing the temperature sensing element at 20°C. FIG 2(A) is an image of the temperature sensing element observed again at 20°C using the high resolution transmission electron microscope after introducing the same into a furnace in the air, heating at 358°C and ~~taking out~~ removal. As shown in FIG 2(A), the top end position of the gallium is made higher than the top end position of the gallium in FIG 1.

Accordingly, the gallium position is made higher despite the measurements at the same ordinary temperature 20°C because the gallium top end part is reacted with ~~an~~-oxygen so as to produce a-gallium oxide, and the produced gallium oxide is firmly bonded with the inner wall of the carbon nano tube so that the gallium oxide layer position at the high temperature is not lowered even after the temperature drop.

This can be confirmed also by the inclusion of ~~the~~-oxygen at the gallium top end part as observed in the energy dispersive X-ray spectroscopy shown in FIG 2(B).

#### <Example 2>

FIG 3 is an image of the same temperature sensing element as ~~the example in~~ Example 1 observed using the high resolution transmission electron microscope after heating to 440°C. FIG 4 is an image thereof observed at 20°C using the same high resolution transmission electron microscope after heating and ~~taking out~~ removal.

From FIG 4, since the gallium oxide layer is bonded firmly with the inner wall of the carbon nano tube, the gallium oxide thin layer position is not changed even after the temperature drop.

It is confirmed that gallium layer (1) of a low density layer is formed in the lower part of the gallium layer, and that the gallium oxide layer is bonded firmly with the inner wall of the carbon nano tube.

As a result of the measurement using the high resolution transmission type electron microscope



accordingly, the height difference of the gallium top ends in FIG 1 and FIG 2(A) was 170 nm.

According to the calculation using the numerical values, the gallium volume  $V_0$  initially at 20°C was  $9.586 \times 10^7 \text{ nm}^3$ , the increased volume  $\Delta V_1$  at the time of heating to 58°C was  $2.333 \times 10^5 \text{ nm}^3$  ( $\Delta V_1/V_0 = 0.24\%$ ), and the volume increase amount  $\Delta V_2$  at  $T_h$ °C was  $2.577 \times 10^6 \text{ nm}^3$ . By substituting these values in the formula  $T_h = 58 + \Delta V_2/a_0 (V_0 + \Delta V_1)$ , (wherein  $a_0$  is the expansion coefficient of the gallium at 58°C [ $0.95 \times 10^{-4}/^\circ\text{C}$ ]),  $T_h = 341^\circ\text{C}$  can be obtained as the calculation value.

Although the value is slightly lower than the real measurement value 358°C, it was confirmed that the temperature can be measured considerably accurately at a high temperature.

As to the difference ~~of~~ in the calculation value and the real measurement value, it is considered that the gallium volume  $V_0$  calculated with the premise that the carbon nano tube inner diameter is increased linearly, is larger than the real gallium volume, and that the gallium density is lowered due to ~~the~~ oxygen diffusion, or the like.

#### Industrial Applicability

According to the present invention, the temperature measurement of the micrometer size environment can be enabled, and furthermore, the temperature measurement ~~in~~ over a wide temperature range of 50 to 500°C can be enabled.

## Abstract

By preliminarily measuring the gallium temperature and the length change of a temperature sensing element comprising a carbon ~~nanotube~~ nanotube having a continuous ~~columnar~~ column of gallium contained therein, then heating the temperature sensing element installed in a subject to ~~have~~ increase the temperature ~~measure thereof in the air, taking out~~ removing the temperature sensing element subjecting to measure the gallium length, and substituting the measured gallium length in ~~the~~ a formula, the temperature is measured accurately ~~in~~ over a wide temperature range in a micrometer size or less environment.